Aerosol-Radiation-Microphysics Interactions

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Motivation



Use WRF-Chem to study local to regional-scale evolution of particulates and their effect on radiation, clouds, and chemistry

A Brief History ...

- First, aerosol-radiation-cloud interactions were coupled to the MOSAIC aerosol model, adapted from those used in a global climate model
- Aerosol-radiation-cloud interactions have been expanded to handle more aerosol models (GOCART, MADE/SORGAM, MAM) and microphysics schemes (Lin, Morrison, Morrison-Gettelman)
- More capabilities are being added and tested, making modules more generic, and trying to follow WRF coding guidelines

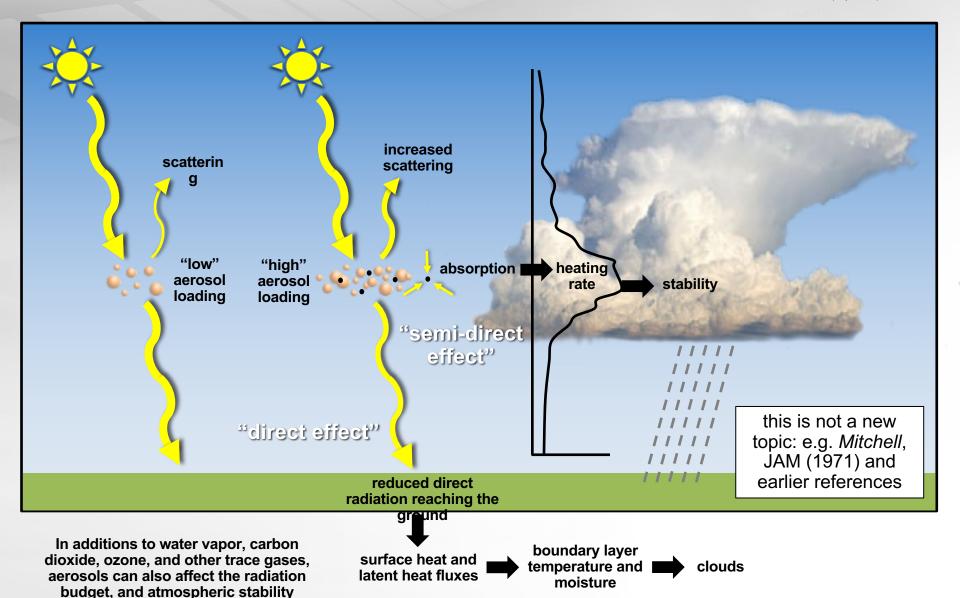
Outline:

- Part 1: Direct Effects
- Part 2: Indirect Effects

Part 1: Aerosol Direct Effects



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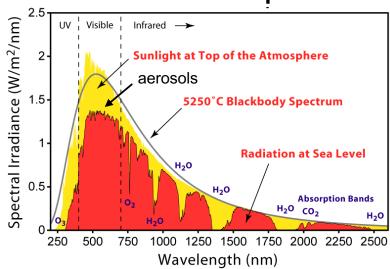
Aerosols in Relation to Radiation Modules



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- Aerosols affect radiation mostly in the visible wavelength region
- In contrast with water vapor, carbon dioxide, and ozone, the <u>temporal and</u> <u>spatial variability</u> of aerosols is much larger and difficult to simulate
 - Episodic Sources: dust, biomass burning, volcanic (potentially large concentrations)
 - More "Continuous" Sources: seasalt, biogenic, anthropogenic (usually smaller concentrations)

Solar Radiation Spectrum



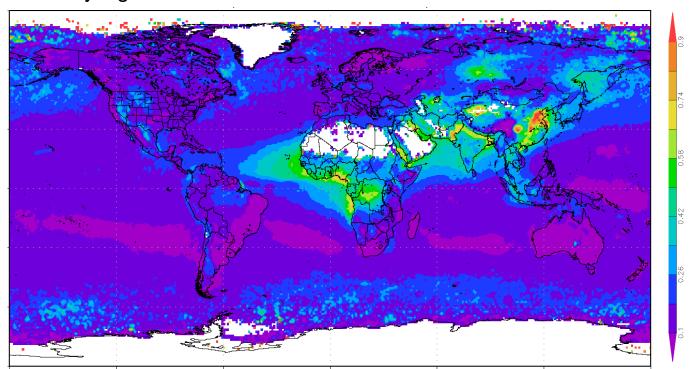
How are aerosol effects accounted for in atmospheric models?

- Ignored no effect of aerosols on radiation
- Use prescribed or climatological aerosol properties that may vary in space and seasonally (not described in this presentation)
- ► Use **prognostic aerosols** (e.g. WRF-Chem)

Aerosol Optical Depth (AOD)



- Extinction coefficient: fractional depletion of radiance per unit path length (km⁻¹) due to scattering and absorption by aerosols
- Aerosol optical depth (AOD) or thickness (AOT): integrated extinction coefficient over a vertical column, I / I_o = e^{-AOD}
 - AOD = 0 no aerosol effect
 - AOD ~ 1 "large"
 - AOD > 1 extremely high aerosol concentrations



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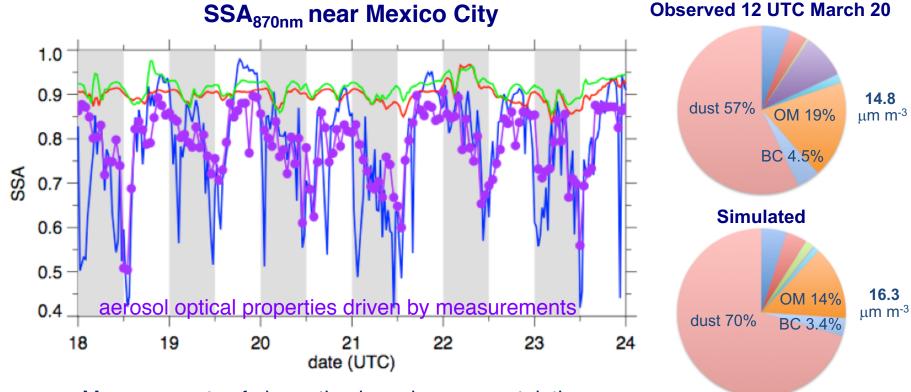


Single Scattering Albedo, ω_{o}



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- SSA is ratio of scattering to extinction efficiency, $\omega_0 = k_s / (k_a + k_s)$
 - SSA = 1 all particle extinction due to scattering
 - SSA = 0 all particle extinction due to absorption (does not happen in reality)
- \blacktriangleright Models simulate AOD_λ "reasonably well", but there are large uncertainties in ω_o



Measurements of absorption have large uncertainties → model-obs comparisons should be done carefully

Asymmetry Factor, g

g from measurements



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- Preferred scattering direction (forward or backward) for the light encountering the aerosol particles
 - Approaches 1 for scattering strongly peaked in the forward direction
 - Approaches -1 for scattering strongly peaked in the backward direction
 - g = 0 means scattering evenly distributed between forward and backward scattering (isotropic scattering such as from small particles)

Depends on both size and composition of aerosols

small particles:
< 1/10 the λ of light

large particles:
> λ of light

Theoretical relationships used to derive

OM

sea-salt

dust

Methodology for Prognostic Aerosols



Generic Aerosol Optical Property Module



AOD, ω_o, and g computed at
 4 wavelengths (300, 400, 600, 1000 nm) for shortwave radiation
 16 wavelengths for longwave radiation

Angstrom exponent used to convert to wavelengths needed by radiation schemes

- Compatible with GOCART, MADE/SORGAM, MOSAIC, and MAM aerosol models as of v3.5
- Compatible with Goddard shortwave scheme and RRTMG shortwave and longwave schemes
- Evaluating aerosol size, number distribution, and composition against measurements is essential before calculating optical properties: If garbage is going into the module, then garbage will come out

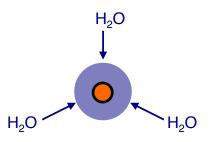
Importance of Aerosol Water



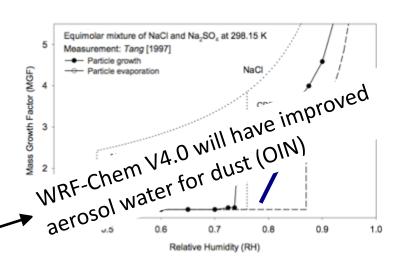
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Aerosol water will have a big impact on optical properties





- Uptake of water by aerosols depends on relative humidity (RH); predictions of RH need to be examined when evaluating aerosol direct radiative effects
- Composition affects water uptake: hydrophobic vs. hydrophillic aerosols
- Aerosols models have different methods of computing aerosol water
 - **GOCART:** Petters and Kreidenweiss (2007)
 - MADE/SORGAM: diagnosed
 - MOSAIC: prognostic specie that accounts for hysteresis effect (currently being updated for OIN species)
 - **MAM:** prognostic specie, Kohler theory



Refractive Indices



- Refractive index of a substance is a dimensionless number that describes how
- light propagates through a medium
- Refractive indices in models based on literature values derived from laboratory experiments, vary with wavelength for some aerosol compositions

Default Values for SW Radiation in WRF (users can change)

	real part		imaginary part
BC =	1.850	+	0.71i (all λ)
OM =	1.450	+	0.00i (all λ)
$SO_4 =$	1.468	+	1.0e-9i (300 nm), small λ dependence
NH ₄ NO ₃	= 1.500	+	0.00i (all λ)
NaCl =	1.510	+	0.866e-6i (300 nm), small λ dependence
dust =	1.550	+	0.003i (all λ), depends on type of dust
$H_2O =$	1.350	+	1.52e-8i (300 nm), small λ dependence
			greater the # more absorption

similar relationships for LW radiation

- On-going research:
 - secondary organic aerosols (SOA) may be absorbing at near-UV range
 - how to handle "brown carbon"

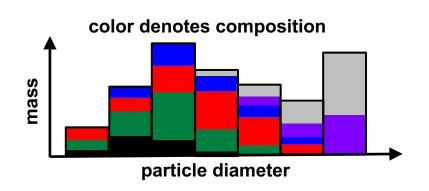
Mixing Rules for Mie Calculations



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Prior to the Mie calculations, refractive indices need to be averaged among the compositions in some way for discrete size ranges of the aerosol size distribution.

All particles within a size range assumed to have the same composition, although relative fraction can differ among size ranges.



Currently three choices in WRF:

Volume Averaging: averaging of refractive indices based on composition



▶ **Shell-Core**: black carbon core and average of other compositions in shell (Ackermann and Toon, 1983; Borhren and Huffman, 1983)



Maxwell-Garnett: small spherical randomly distributed black carbon cores in particle (Borhren and Huffman, 1983)

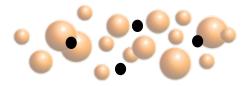


Mie Calculations

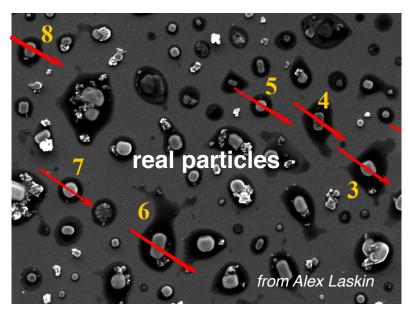


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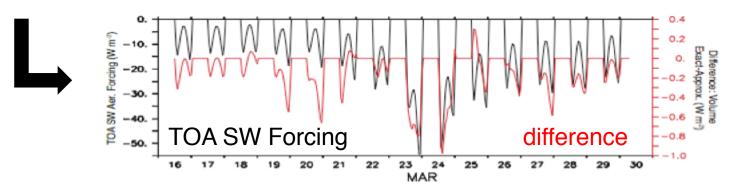
The Mie solution to Maxwell's equations describes the scattering of radiation by a sphere, used to obtain AOD_{λ} , ω_{o} , and g



- Aerosols are rarely spheres; however, aged aerosols become more "sphere-like"
- Several "standard" codes available and one is included in WRF
- Mie codes can be computationally expensive, so an approximate version (Ghan et al. JGR, 2001) is also available



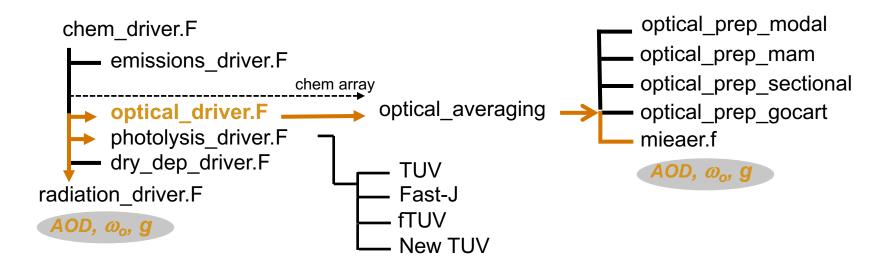
other codes available to handle more complex morphology, but not clear if it is really necessary



Coding Structure



Generic Aerosol Optical Properties Module for WRF-Chem



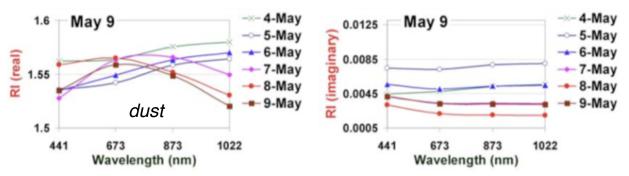
Example of making the code more generic and interoperable: optical property is calculated in one routine rather than in each aerosol model

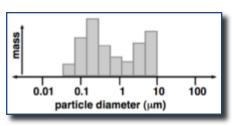
Assumptions of Optical Property Module

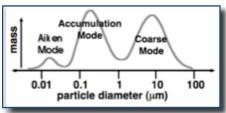


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- Interfaces with GOCART, MADE/SORGAM, MAM, and MOSAIC, but linking to other aerosol models should be relatively easy
- ► Sectional (MOSAIC): tested only with 4 and 8 size bins should work if additional size bins are specified
- Modal (MADE/SORGAM, MAM): maps the used size modes into 8 sections
- ▶ **Bulk** (GOCART): converts bulk mass into assumed distribution, then divides mass into 8 sections
- Note: Refractive indices may need updating
 - Range of values reported in the literature
 - Wavelength dependence of refractive indices for some species





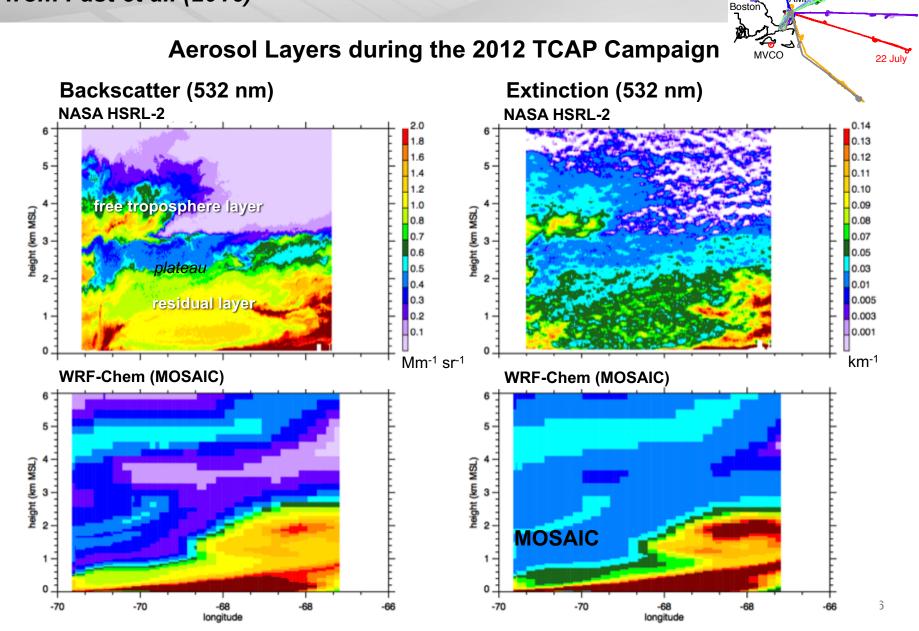


Dust refractive indices for SW constant by default – need to modify code to turn on

from Prasad and Singh, JGR, 2007

Example: Evaluating Extinction Profiles

from Fast et al. (2016)



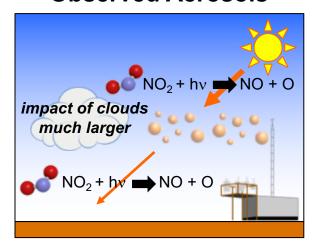
AME

Impact of Aerosols on Chemistry

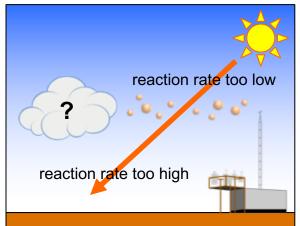


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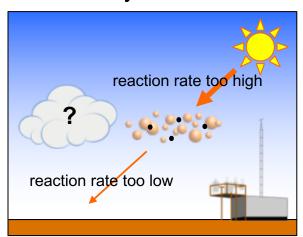
Observed Aerosols

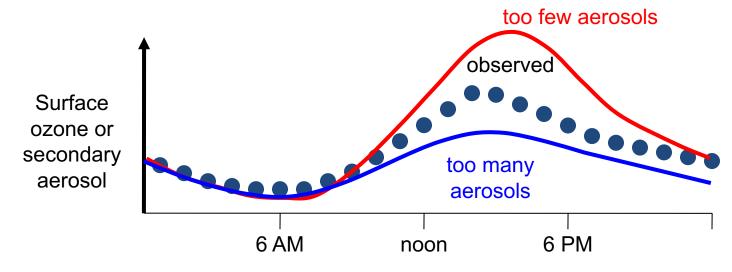


Simulated:
Too Few or Too Thin



Simulated: Too Many or Too Thick



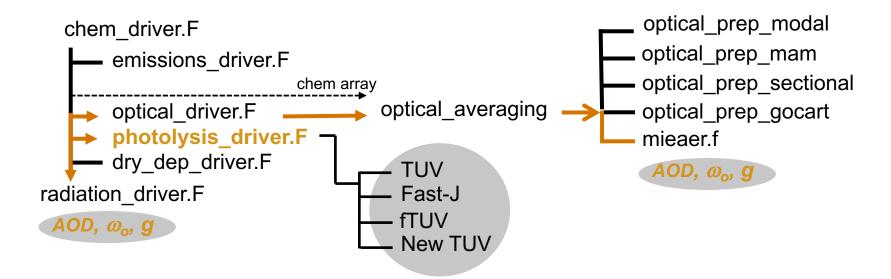


errors could impact simulated concentrations the next day

How Aerosols Affect Photolysis Rates



Aerosols Photolysis Rates Photochemistry
but clouds (if present) will have a bigger impact on photolysis rates than aerosols



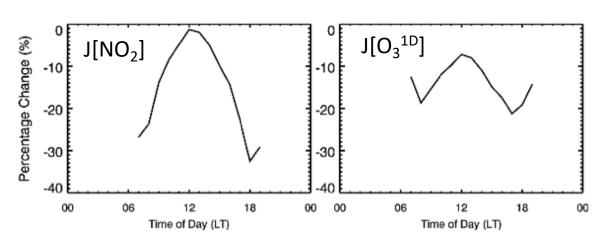
- Fast-J: uses AOD, ω_o, and g computed by module_optical_averaging.F
- FTUV: uses AOD, ω_o, and g computed by module_optical_averaging.F
- Updated TUV: uses AOD, ω_o, and g computed by module_optical_averaging.F

Example: Impact of Aerosols on Photolysis

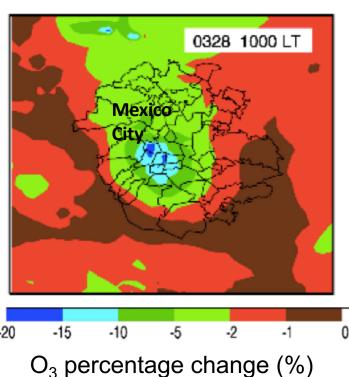
from Li et al. ACP (2011)



Aerosol effects on surface photolysis and ozone in Mexico City



- Decrease in J[NO₂] and J[O₃^{1D}] values during the day
- Decrease in surface ozone concentrations by 5-20% within the Mexico City

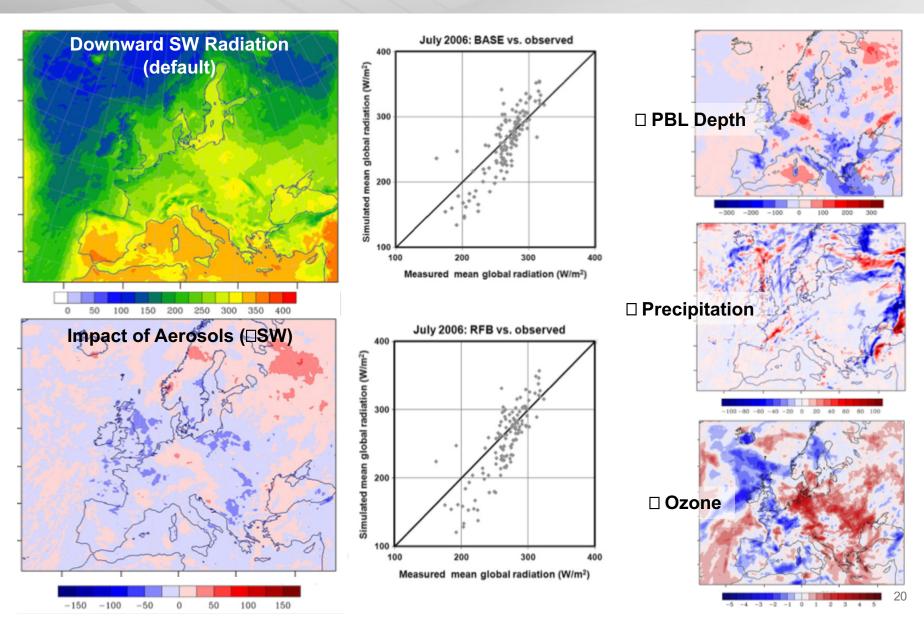


Example: Impact of Aerosols over Europe

from Forkel et al. ACP (2012)



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Settings in namelist.input



Important Parameters:

- ra_sw_physics = 2 aerosols affect radiation computed by Goddard scheme
- ra_sw_physics = 4
 ra_lw_physics = 4
 aerosols affect radiation computed by RRTMG scheme
- aer ra feedback = 1, turns on aerosol radiation feedback
- aer_op_opt = > 0, define the mixing rule for Mie calculations
- Works similarly for GOCART, MADE/SORGAM, MAM, and MOSAIC options

Aerosol-Radiation Interactions:

- Simulations with aer_ra_feedback = ON or OFF can be used to quantify direct effects, but differences in clouds complicates interpretation
 - Useful to add code that computes radiation with and without aerosols and with and without clouds (either directly in the code or computed off-line)
 - Or work with small perturbations in aerosol fields

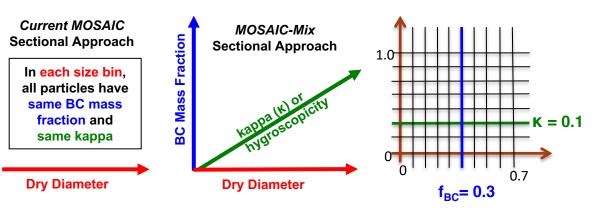
Future Capabilities



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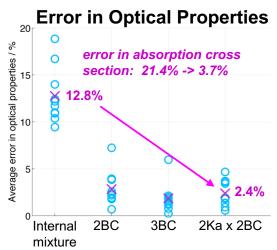
Research – Possibly in Upcoming Releases of WRF:

- Different refractive indices organic aerosol components
- More computationally efficient Mie calculations
- More detailed treatment of optical properties of organic aerosols including treatment for "brown carbon"
- Code to handle aerosol model with external mixtures



Ching et al., 2016, JGR also see Matsui et al., 2014, ACP

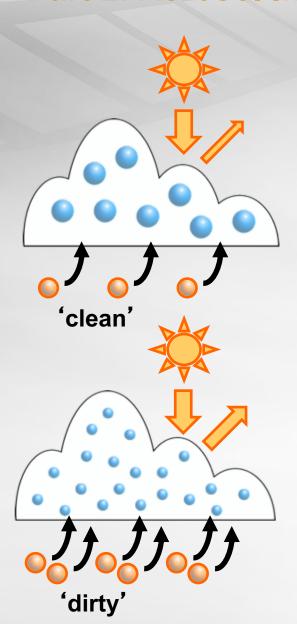
identify optimal bin boundaries compared to explicit calculations



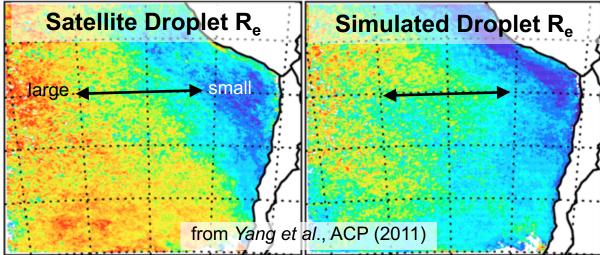
Part 2: Aerosol Indirect Effects



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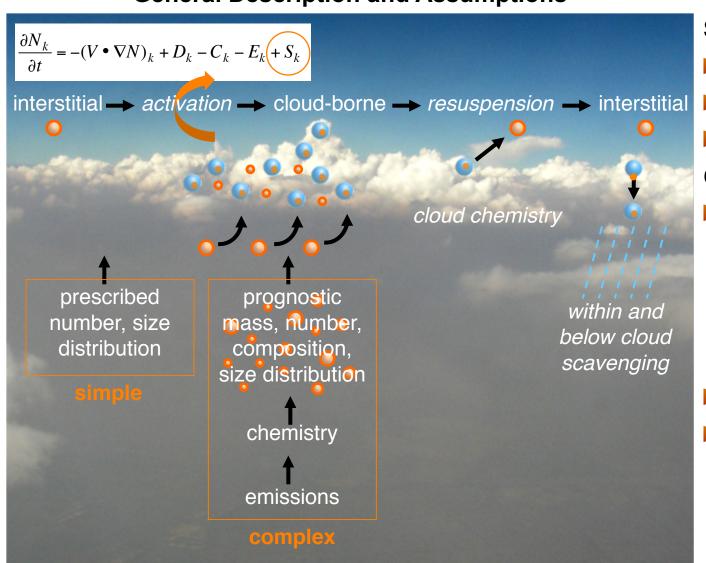
The number of activated aerosols affects the cloud drop size distribution, and consequently cloud albedo and radiation budget

Aerosol-Cloud Interactions in grid-scale clouds



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General Description and Assumptions



Simple:

- chem_opt=0
- progn = 1
- naer = specified

Complex:

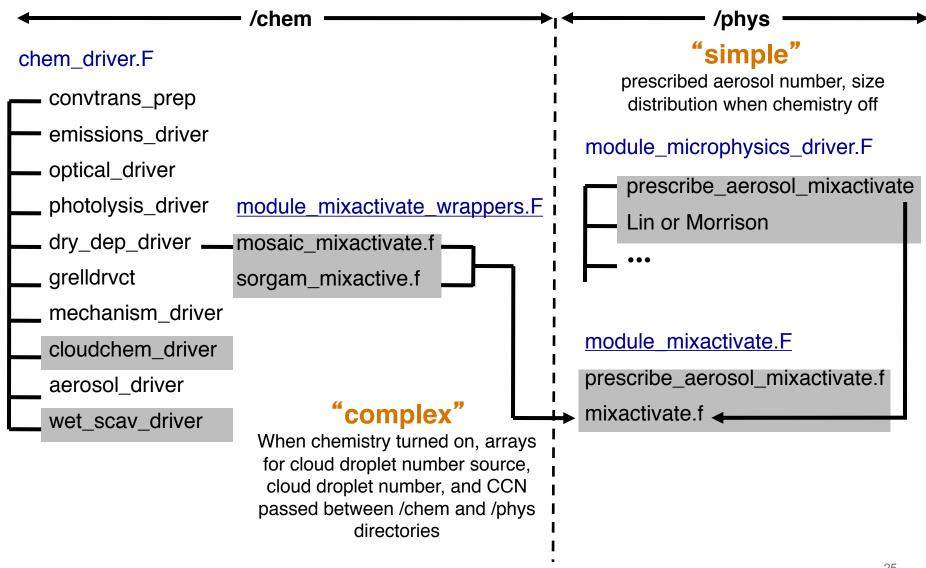
- chem_opt =
 9-12, 32, 34, 35,
 41-43, 132, 202,
 203, 503, 504,
 601, 611
- progn = 1
- naer = ignored

coupled to 2 microphysics schemes: Lin and Morrison

Flow Chart



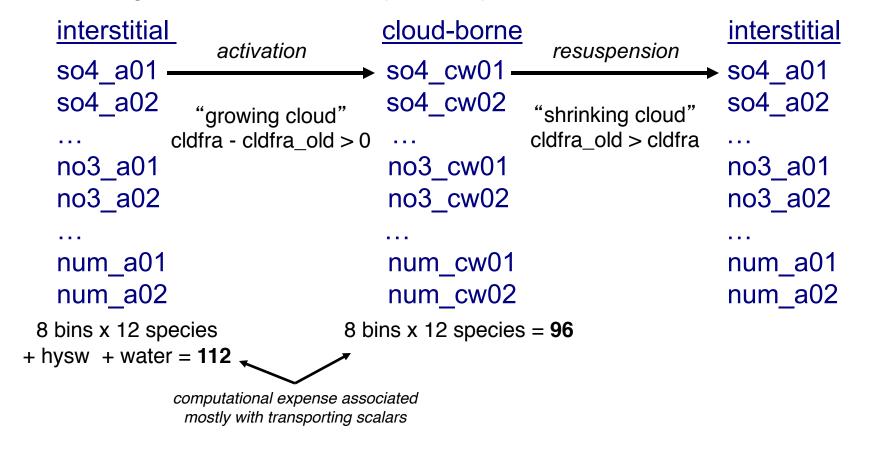
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Aerosol Species



 Interstitial and cloud-borne aerosol particles treated explicitly, nearly doubling the number of transported species

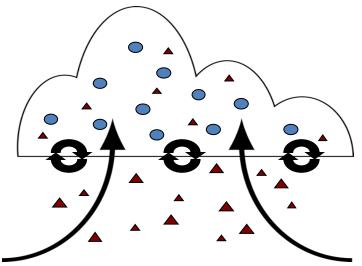


▶ Similar for MADE/SORGAM: so4aj → so4cwj → so4a

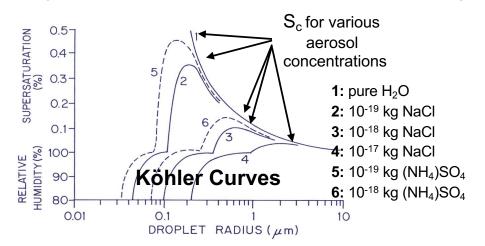
Activation



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Aerosols activated when the environmental supersaturation in the air "entering cloud", S_{max} > aerosols critical supersaturation, S_c



Activate.f computes activation fraction for mass and number for each bin/mode. Inputs include mean vertical velocity, *wbar*, and σ of the turbulent velocity spectrum, *sigw*.

Note: *sigw* based on *exch_h*, but some PBL options (ACM) do not have *exch_h* passed out of the subroutine. Minimum *exch_h* set to 0.2 m s⁻¹ since predicted values may be too low in free atmosphere.

For each vertical velocity, peak S_{max} depends on aerosol size and composition [Abdul Razzak and Ghan, 2000, 2002]. Activation fraction based distribution of S_c of the bin/mode - simply a fraction of aerosol mass or number in the bin/mode having $S_c < S_{max}$

Hygroscopicity



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- Hygroscopic properties depend on particulate composition:

 - hygro_no3_aer = 0.5
 - hygro_nh4_aer = 0.5
 - hygro_oc_aer = 0.14 (some OC may be hygrophilic subject of research)
 - hygro_bc_aer = 1.0e-6 hydrophobic ------
 - hygro_oin_aer = 0.14.
 - hygro_ca_aer = 0.1 --->------
 - hygro_co3_aer = 0.1
 - hygro msa aer = 0.58
 - hygro_cl_aer = 1.16
 - hygro_na_aer = 1.16

hydrophilic -----



 CO_3





- Activation depends on volume weighted bulk hygroscopicity, prior to call to mixactivate.f in module_mixactivate_wrappers.F
 - Coating not taken into account
- For chem_opt = 0 and nprog = 1, hygroscopicity set to 0.5

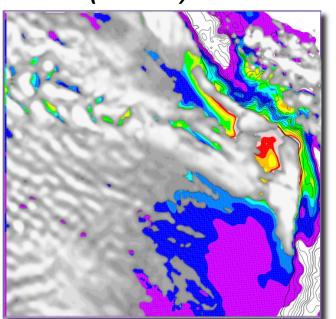
Cloud Condensation Nuclei



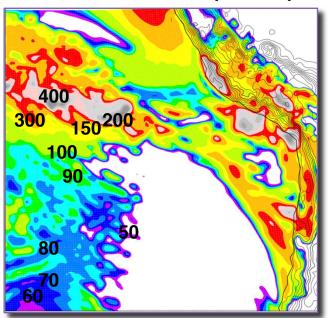
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- CCN: number concentration of aerosols activated at a specified supersaturation
- Diagnostic quantity, varies in space and time (can be measured)
- Computed in module_mixactivate.F at 6 super-saturations (.02, .05, .1, .2, .5, and 1%) that correspond to CCN1, CCN2, CCN3, CCN4, CCN5, CCN6 in Registry

AOD (600 nm) and COD



CCN at 0.1% SS (# cm⁻³)



example from VOCALS-Rex: southeastern Pacific marine stratocumulus

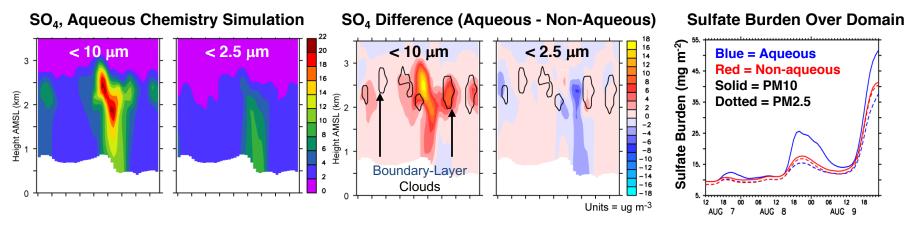
Aqueous Chemistry



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- Bulk cloud-chemistry module of Fahey and Pandis (2001) compatible with MOSAIC and MADE/SORGAM (cloudchem_driver.F)
- Chemistry in cloud drops, but not rain drops
- Oxidation of S(IV) by H₂O₂, O₃, trace metals, and radical species, as well as non-reactive uptake of HNO₃, HCl, NH₃, and other trace gases (aqueous organic chemistry being developed)
- Bulk mass changes partitioned among cloud-borne aerosol size bins, followed by transfer of mass & number between bins due to growth

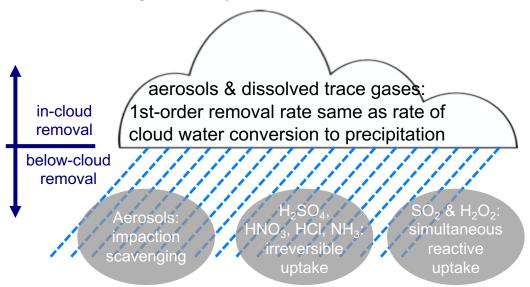
Vertical Cross-Section Though Power Plant SO₂ Plume





 Cloud-borne aerosols and trace gases are collected by both grid-scale and convective precipitation (rain, snow, graupel)

cloud-borne aerosols are explicit, while the fraction of trace gas that is dissolved in cloud water is calculated in the cloud chemistry module



- scavenged aerosols and gases instantly removed Easter et al. (2004); aerosols are not resuspended by evaporating rain
- In MOZART based packages, the washout of trace gases is based on Neu and Prather (2012), and updated solubility coefficients are used for organic gases → WRF-Chem V4.0 adds a HLC table to run directory

Henry's Law Constants Table



- File: HLC.TBL contains Henry's Law and Dissociation coefficients
- ► Henry's Law Constants updated to JPL (2015) recommendations and Sander (2015, ACP) data archive; Some HLC values are estimated based on functional group structure of compound
- ► MOZART chemistry options use this table
 - Chem_opt = 111, 112, 201, 202 and the new MOZCART-T1 option
- Three routines use this table
 - module_mozcart_wetscav.F → wet deposition of trace gases
 - module dep simple.F → dry deposition of trace gases
 - module_ctrans_grell.F → convective wet deposition of trace gases
- ► These three processes use the same Henry's Law constants (which was not the case in earlier versions of WRF-Chem)
- ► The table is not connected to the aqueous chemistry modules

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- Converted Lin et al. microphysics scheme to a two-moment treatment (mass & number), in addition to adding impact of aerosols on droplet #
- Morrison microphysics is a two-moment treatment, so only needed to add code to include the impact of aerosols on droplet #

$$\frac{\partial N_k}{\partial t} = -(V \bullet \nabla N)_k + D_k - C_k - E_k + S_k$$

$$\text{qndrop} \longrightarrow N_k \text{ .grid cell mean droplet number mixing ratio in layer } k$$

$$D_k \text{ - vertical diffusion}$$

$$C_k \text{ - droplet loss due to collision/coalescence & collection}$$

$$E_k \text{ - droplet loss due to evaporation}$$

$$S_k \text{ - droplet source due to nucleation (determined in mixactivate.f)}$$

- cloud droplet number source determined by aerosol activation (for meteorology-only runs a prescribed aerosol size distribution is used)
- droplet number and cloud water mixing ratio used to compute effective cloud-particle size for the cloud optical depth in Goddard or RRTMG shortwave radiation scheme (ra sw physics = 2 or 4)

Example: Marine Stratocumulus

from Yang et al. ACP (2011)

interactions



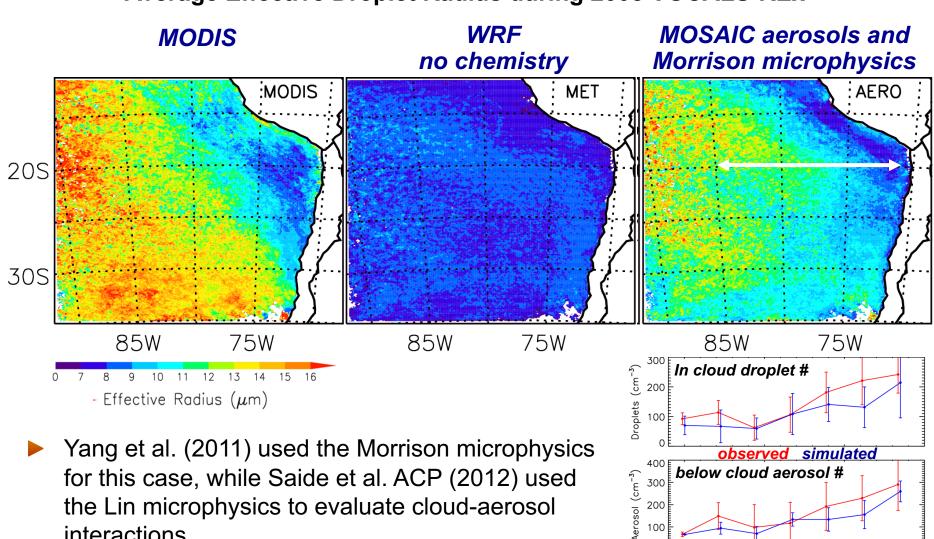
80W

85W

75W

70W

Average Effective Droplet Radius during 2008 VOCALS-REx

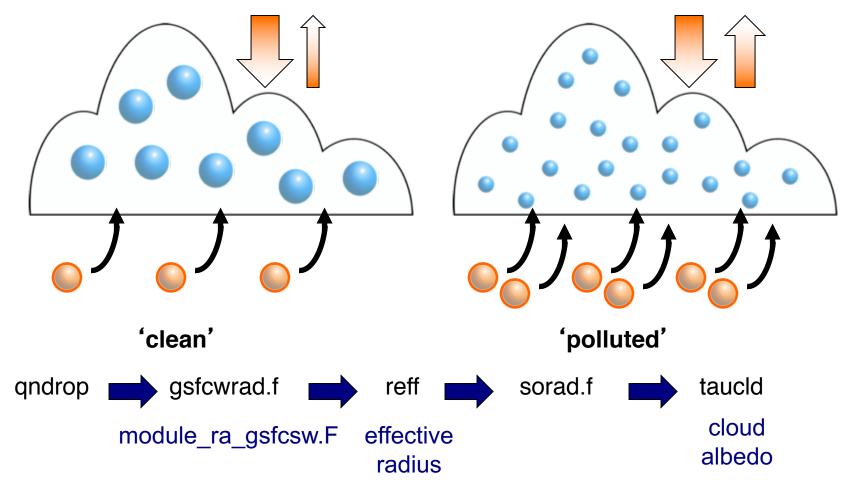


First Indirect Effect



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Influence of cloud optical depth through impact on effective radius, with no change in water content of cloud



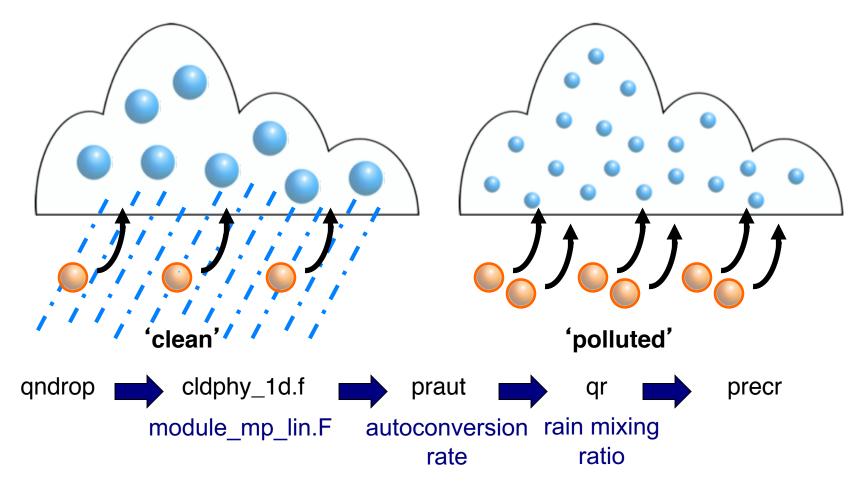
(subroutines for Goddard scheme)

Second Indirect Effect



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Influence of cloud optical depth through influence of droplet number on mean droplet size and hence initiation of precipitation



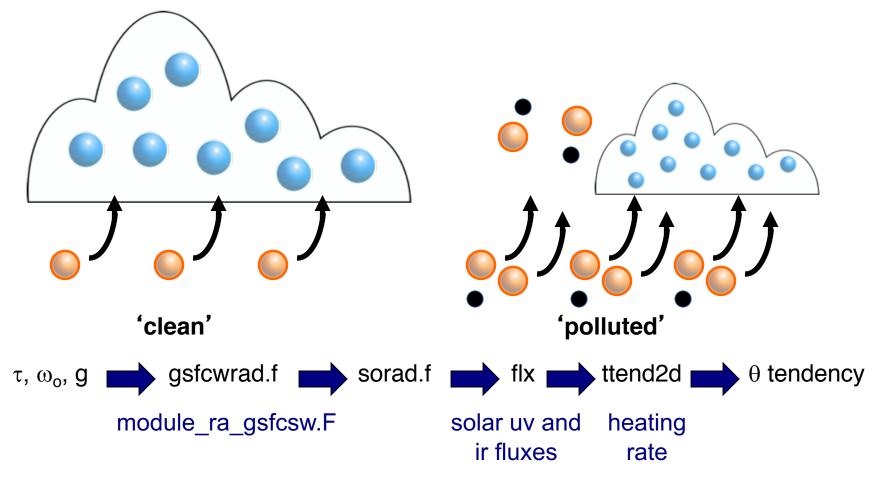
(subroutines for Goddard scheme)

Semi-Direct Effect



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Influence of aerosol absorption of sunlight on cloud liquid water and hence cloud optical depth



(subroutines for Goddard scheme)

Settings in namelist.input



Cloud-Aerosol Interactions for Lin and Morrison Microphysics

- mp_physics = 2, 10
- progn = 1, turns on prognostic cloud droplet number

Simple:

- chem_opt = 0
- naer = specified value

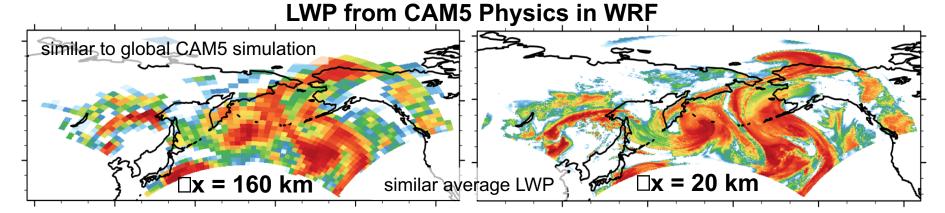
Complex:

- chem_opt = 9, 10, 32, 34, 202, 203, 601, 602 cloud-phase for MOSAIC = 11, 12, 35, 41-43, 132 for MADE/SORGAM = 503, 504 for MAM
- cldchem_onoff = 1, turns on cloud chemistry
- wetscav_onoff = 1, turns on wet scavenging

CAM5 Physics is Different (1)



- Cloud-Aerosol Interactions for Morrison and Gettelman microphysics handled separately, because
 - CAM5 physics kept as same as possible as in the CESM climate model



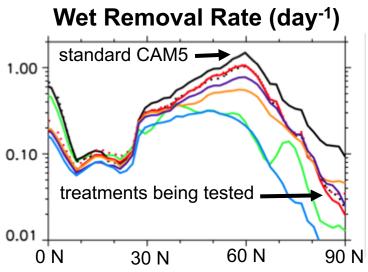
- Entire CAM5 physics suite must be used when simulating cloud-aerosol interactions in the Morrison and Gettelman microphysics scheme
 - mp_physics=19, cu_physics=7, shcu=physics=2, bl_pbl_physics=9 chem_opt=503, cam_mam_mode=3, CAM_MP_MAM_cpled='true'
- /phys/module_mixactivate.F is not used (activation is done elsewhere), but is conceptually similar to how it is handled in WRF for other models

CAM5 Physics is Different (2)



Proudly Operated by Battelle Since 1965

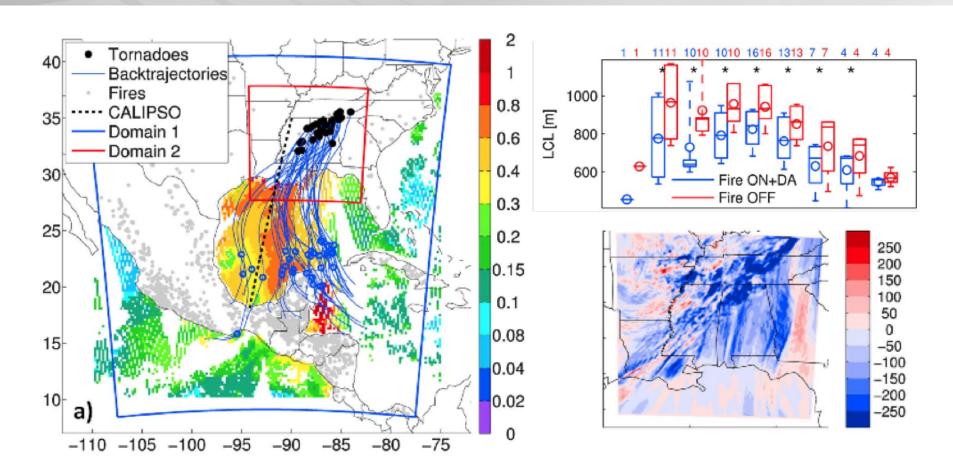
- Morrison and Gettelman microphysics includes treatment of heterogeneous freezing on mineral dust
 - But, there are no ice-borne aerosols
 - Coupling of prognostic aerosols to ice nuclei (IN) not included for other microphysics scheme; the effect of aerosols on cloud droplets will affect ice processes indirectly however
- module_wetscav_driver.F modified to handle MAM aerosols
 - See Wang et al. GMD (2013) for a discussion on wet removal and its uncertainties
- CAM5 physics in WRF is described in (Ma et al., 2014 GMD) paper.



Example: Smoke and Tornado Severity



from Saide et al. GRL (2015)



Inclusion of smoke to an environment already conducive to severe thunderstorm development can increase the likelihood of significant tornado occurrence



Care Must be Taken in Quantifying Indirect Effects!

Indirect Effects:

- Comparing runs with chem_opt = 8 (without cloud-borne aerosols) with chem_opt = 10 (with cloud-borne aerosols) for MOSAIC coupled to Lin microphysics does not quantify the indirect effect
 - since the autoconversion scheme used in the Lin microphysics scheme will be different
 - Need to determine a prescribed aerosol scenario to compare with chem_opt =10 see Gustafson et al., GRL, (2007)
 - An approach used with GCMs is to output "dirty-cloudy", "dirty-clear", "clean-cloudy", and "clean-cloudy" radiation from the same run

Indirect Effects Usage:

- In addition to Abdul-Razaak and Ghan (2000, 2002), other schemes have been used to compute aerosol activation (Foutoukis and Nenes, 2005)
- Works with microphysics only not cumulus parameterizations so users must be aware of issues associated with spatial scale

New Option for Parameterized Clouds

Modifications to Kain-Fritsch Cumulus

- Used Cumulus Potential (CuP) approach to improve the simulation of shallow cumuli (Berg et al., MWR, 2013)
- Cloud fraction of both active and passive clouds

New WRF-Chem chemistry package coupled with MOSAIC aerosol –

WRF-Chem Top Level

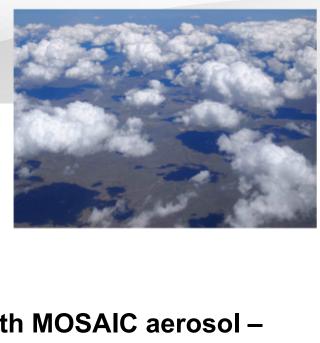
Surface

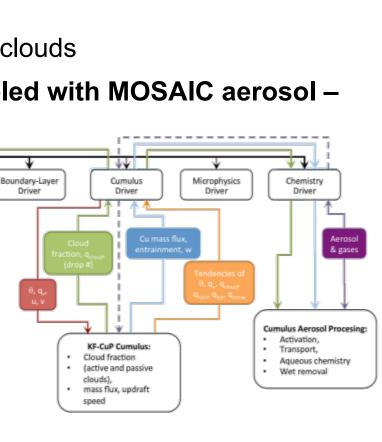
see Berg et al., GMD, 2015

Vertical transport of gases and aerosols

Aerosol activation / resuspension

- Aqueous chemistry (gas SO₂ + S(IV) in cloud water)
- Wet removal of gases and aerosols
- Feedbacks to radiation, precipitation, and cloud lifecycle not yet included







Processes Under Development:

- ACI in Thompson cloud microphysics (not fully coupled to WRF-Chem)
- ACI in spectral bin cloud microphysics with MOSAIC (Gao et al., JAMES, 2016)
- Other treatments are likely being developed by WRF-Chem users that are not known until they are published
- Aqueous-phase organic chemistry affecting SOA concentrations
- Resuspension of aerosols from evaporating rain
- Secondary activation (Yang et al. JGR, 2015)
- Ice-borne MOSAIC aerosols and IN treatment

→Still many processes to better represent

For more information and updates:

- PNNL modules: www.pnl.gov/atmospheric/research/wrf-chem
- See web page for list of papers on aerosol-cloud interactions (ACI)